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## PATENT ABSTRACTS OF JAPAN

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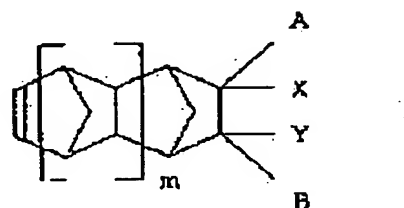
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(22)Date of filing : 11.12.1991

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OSHIMA NOBORU**(54) PRODUCTION OF HYDROGENATED POLYMER****(57)Abstract:**

**PURPOSE:** To obtain a hydrogenated polymer useful as a tackifier for adhesive, etc., having excellent thermal stability, tack, bond strength and retention power by polymerizing a mixture of a cyclopentadiene-based monomer and a specific norbornene-based compound and hydrogenating the polymer.

**CONSTITUTION:** (A) 0-90wt.% cyclopentadiene-based monomer is thermally polymerized with 10-100wt.%, preferably 20-100wt.%, more preferably 40-100wt.% norbornene-based compound of formula I [A and B are H or 1-10C hydrocarbon group; X and Y are H or monofunctional organic group and at least one of X and Y is H and polar group except hydrocarbon group, preferably group of formula II (R1 is 1-20C hydrocarbon group; (n) is preferably 0); (m) is 0-2] preferably in the absence of a radical initiator at 200-300° C and (B) the polymer is hydrogenated by using a catalyst (preferably palladium supported on a silica or magnesia carrier) under normal pressure to 300 atmospheric pressure in a hydrogen gas atmosphere at 0-300° C to produce the hydrogenated polymer.

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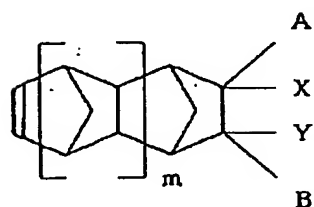
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## CLAIMS

## [Claim(s)]

[Claim 1] The manufacture approach of the hydrogenation polymer which carries out the polymerization of the 10 - 100 % of the weight of the norbornene system compounds shown by 0 - 90 % of the weight of cyclopentadiene system monomers, and the general formula (I), and is characterized by subsequently hydrogenating the obtained polymer under existence of a hydrogenation catalyst.

## [Formula 1]



[-- X and Y of A and B are a hydrogen atom or the organic radical of monovalence among a formula about a hydrogen atom or the hydrocarbon group of carbon numbers 1-10, at least one of X and the Y shows the radical which has polarities other than a hydrogen atom and a hydrocarbon group, and m is 0-2.]

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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the manufacture approach of a new hydrogenation polymer.

[0002]

[Description of the Prior Art] Generally hydrogenation polymers, such as a hydride of the poly cyclopentadiene, are used as a tackifier for hot melt mold adhesives which makes a base polymer the polymer which hydrogenated the diene section of a styrene butadiene block copolymer, a styrene isoprene block copolymer, and these polymers.

[0003]

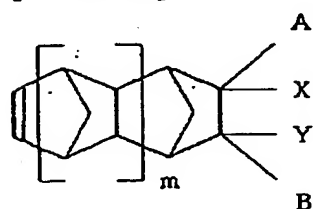
[Problem(s) to be Solved by the Invention] However, compared with organic solvent mold adhesive tape, the adhesive strength and the low-temperature tack at the time of an elevated temperature were insufficient for the hot melt mold binder which made the conventional hydrogenation polymer the tackifier, and development of the tackifier from which the hot melt mold binder of high performance is obtained more was desired.

[0004]

[Means for Solving the Problem] The purpose of this invention is to offer the manufacturing method of the hydrogenation polymer excellent in the hue and the adhesion property. That is, this invention carries out the polymerization of the 10 - 100 % of the weight of the norbornene system compounds shown by 0 - 90 % of the weight of cyclopentadiene system monomers, and the general formula (I), and offers the manufacture approach of the hydrogenation polymer characterized by subsequently hydrogenating the obtained polymer under existence of a hydrogenation catalyst.

[0005]

[Formula 2]



[-- for the inside A and B of a formula, it is a hydrogen atom or the hydrocarbon group of carbon numbers 1-10, and X and Y are a hydrogen atom or the organic radical of monovalence, at least one of X and the Y shows the radical which has polarities other than a hydrogen atom and a hydrocarbon group, and m is 0-2.] Hereafter, the component of this invention is explained in full detail. As a cyclopentadiene system monomer used in this invention, lower order Diels-Alder adducts, such as \*\*\*\*\* of those \*\*\*\*\*, \*\*\*\*\*, \*\*\*\*\* and a cyclopentadiene (alkylation), and dienes, such as an isoprene and piperylene, etc. can be mentioned to the dimer of an others, a cyclopentadiene, or its alkylation object, a trimer, a tetramer, and a pan. [ cyclopentadiene / like a cyclopentadiene, a methyl cyclopentadiene, an ethyl cyclopentadiene,

and a dimethyl cyclopentadiene / low-grade alkylation ]

[0006] Moreover, a part of cyclopentadiene system monomer can also be replaced with the ester of alpha, such as dienes, such as styrene system hydrocarbon compounds, such as an indene, a methyl indene, cumarone, etc. and styrene, alpha-methyl styrene, a butadiene, and an isoprene, a methyl acrylate, and a methyl methacrylate, and beta unsaturated carboxylic acid etc. As a norbornene system compound shown by the general formula (I) which can be used in this invention A 5-methoxycarbonyl bicyclo [2.2.1] hept-2-en, A 5-butoxycarbonyl bicyclo [2.2.1] hept-2-en, A 5-cyclohexyloxy carbonyl bicyclo [2.2.1] hept-2-en, A 5-(4'-t-butyl cyclohexyloxy) carbonyl bicyclo [2.2.1] hept-2-en, 5 - Methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en, 5 - Methyl-5-(1-methyl propoxy) carbonyl bicyclo [2.2.1] hept-2-en, A 5-methyl-5-phenoxy carbonyl bicyclo [2.2.1] hept-2-en, 8-methoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 4.4.0.12, 5.17, 8-(1-methyl propoxy) carbonyl tetracyclo [10]-3-dodecen, 8-phenoxy carbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3 - Dodecen, 8-methyl-8-methoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-ethoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-isopropoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-cyclohexyloxy carbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-(4'-t-butyl cyclohexyloxy) carbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, and 8-cyano tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, etc. can be mentioned.

[0007] The hydrogenation polymers obtained as a radical which has a polarity in the norbornene system compound expressed with the general formula (I) used in this invention are an elevated temperature, high adhesive strength, and the point of having low hygroscopicity, the radical expressed with formula-(CH<sub>2</sub>)<sub>n</sub>COOR<sub>1</sub> is desirable, and this radical is desirable because the hygroscopicity of the hydrogenation polymer with which it is acquired that it is one per norbornene compound 1 molecule expressed with (a general formula I) becomes low. Moreover, n among the radicals expressed with formula-(CH<sub>2</sub>)<sub>n</sub>COOR<sub>1</sub> That it is desirable since the thermal stability of the hydrogenation polymer with which the thing which has a small value is obtained becomes high, and it is n= 0 usually The hydrogenation polymer obtained when compounding a monomer is desirable at the point that a good property is acquired. the hygroscopicity of a hydrogenation polymer becomes small so that a carbon number increases, although R<sub>1</sub> is the hydrocarbon group of carbon numbers 1-20, but since softening temperature and thermal stability fall, the point of balance with the softening temperature of a hydrogenation polymer and the hygroscopicity which are acquired to R<sub>1</sub> is the chain-like hydrocarbon group of carbon numbers 1-4, or the cyclic-hydrocarbon radical of carbon numbers 5-7 -- it is desirable.

[0008] Furthermore, it is desirable in order that that by which the hydrocarbon group of carbon numbers 1-10, especially the methyl group are permuted by the carbon atom which the radical expressed with formula-(CH<sub>2</sub>)<sub>n</sub>COOR<sub>1</sub> combined may reduce hygroscopicity, without reducing the softening temperature of the hydrogenation polymer obtained. The ratio of the cyclopentadiene system monomer in monomer mixture is 0 - 60% of the weight of the range more preferably zero to 80% of the weight zero to 90% of the weight. If this ratio exceeds 90 % of the weight, when it uses as a tackifier of a hot melt constituent, the hydride of the polymer obtained will not be able to give sufficient cold resistance, and will be inferior also in the adhesive property at the time of an elevated temperature. The ratio of the norbornene system compound shown by the general formula in monomer mixture (I) is 10 - 100 % of the weight, and is 40 - 100 % of the weight more preferably 20 to 100% of the weight. If this ratio is less than 10 % of the weight, when that hydride is used for a hot melt constituent, it becomes impossible to demonstrate that unique adhesiveness, and is not desirable.

[0009] In this invention, the polymerization with the norbornene system compound shown by the cyclopentadiene system monomer and the general formula (I) can be performed under existence of a solvent or un-existing by various kinds of approaches, such as cationic polymerization, anionic polymerization, ion coordination polymerization, a radical polymerization, and non-catalyst thermal polymerization. In a cationic polymerization catalyst, for example, a cationic polymerization nature catalyst No. [ 10 ] given [ 16th-volume ] in the petrochemical meeting magazine 865-867th page (1973), Specifically AlCl<sub>3</sub>, AlBr<sub>3</sub>, BF<sub>3</sub>, SnCl<sub>4</sub>, SbCl<sub>5</sub>, FeCl<sub>3</sub>, AlRCl<sub>2</sub> (R: alkyl group), There is AlEt<sub>3</sub>-H<sub>2</sub>O, CCl<sub>3</sub>COOH, and H<sub>2</sub>SO<sub>4</sub> etc., and in these, the yield of a

polymer is high, a hue is good, and Lewis acid, especially  $AlCl_3$ ,  $AlBr_3$ , etc. are excellent in the point that the high thing of softening temperature is obtained. Moreover, complexes, such as these Lewis acid and alcohol, ester, the ether, and alkyl halide, can also be used suitably.

[0010] For an ion coordination polymerization catalyst, on a Ziegler type catalyst No. [ 8 ] given [ for example, / 16th-volume ] in the petrochemical meeting magazine 699-704th page (1973), and a concrete target The halide of Ti, V, Cr, and Zr, beta-diketone salt, an alcoholate, etc.  $AlR_3$ ,  $AlR_nX_{3-n}$ ,  $LiR$ , a  $MgRX$  [R:alkyl group, X: There is combination with a halogen atom, n:1 or 2],  $LiH$ , etc., and in these, since reactivity is high, the halide of Ti or V and its combination of alkylaluminum are desirable. According to radical polymerization catalyst No. [ 10 ] given [ for example, / 16th-volume ] in the petrochemical meeting magazine 867-871st page (1973), and the polymerization temperature to which specifically has redox system catalysts, such as azo compounds, such as peroxides, such as benzoyl peroxide and tertiary butyl high dollar peroxide, and azo-isobutyro-dinitrile, the hydrogen-peroxide-1st iron compound, and potassium persulfate, and these radical polymerization catalyst was set, it is suitably chosen as radical polymerization catalyst from these.

[0011] Although it can apply if a lot of [ the so-called metathesis ring opening polymerization ] molecular weight modifiers (for example, 1-hexane etc.) are not added in the polymerization in this invention, and there is nothing that there is nothing, since it is generally easy to give the amount polymer of macromolecules, it is not desirable. In this invention, especially the thing done for thermal polymerization at 200-300 degrees C is desirable under un-existing [ of a radical initiator ] among these polymerizations. In this case, 230-280 degrees C of polymerization temperature can be preferably acquired by carrying out thermal polymerization at 240-270 degrees C more preferably. In this case, polymerization time amount is 0.5 - 20 hours. The molecular weight of the polymer obtained is polystyrene conversion, 300-3000 are desirable, it is desirable that the thing of 5000 or more molecular weight does not contain 5% of the weight or more especially, and the thing of 5000 or more molecular weight is 2 or less % of the weight more preferably. In this invention, the hydrogenation reaction of this polymer is performed by the usual approach. What is used for the hydrogenation reaction of the usual olefin nature compound can be used for the catalyst used at this hydrogenation reaction.

[0012] For example, as a heterogeneous catalyst, the solid-state catalyst which made support, such as carbon, a silica, an alumina, and a titania, support catalyst matter, such as palladium, platinum, a ruthenium, a rhodium, and nickel, is mentioned. The catalyst which supported especially palladium given in JP,3-174406,A to silica magnesia support is activity and a life. It is desirable from points, such as cost, and when a heterogeneous catalyst is used, it can react to a suspension floor and the solid-state floor. Moreover, the catalyst which combined organic lithiums, such as organic aluminum, such as organic solvent soluble nickel [ , such as naphthenic-acid nickel, titanocene dichloride, and cobalt acetylacetonate, ], cobalt, titanium, vanadium compound and triethylaluminum, triisobutylaluminum, and diethyl aluminum mono-chloride, or butyl lithium, as a homogeneous catalyst can be used, and noble-metals complex compound catalysts, such as a chloro tris (triphenyl phosphine) rhodium, can also be used. a hydrogenation reaction -- ordinary pressure - 300 atmospheric pressure -- desirable -- the bottom of the hydrogen gas ambient atmosphere of three to 150 atmospheric pressure, and temperature -- 0-degreeC-300-degreeC -- it can carry out by 20-degreeC-250-degreeC preferably.

[0013] According to this invention, the hydrogenation polymer which has the outstanding hue, i.e., an one or less Gardner color standards [ which were specified to JIS K-5400 ] hue, and has the softening temperature specified to JIS K-2531 [ 60-200-degree C ] is obtained. The hydrogenation polymer obtained by this invention is meltable to aliphatic hydrocarbon, such as a pentane, a hexane, benzene, toluene, a xylene, chloroform, and a carbon tetrachloride, aromatic hydrocarbon, or halogenated hydrocarbon, and is a polymer-like polymer in which low melt viscosity is shown. The hydrogenation polymer obtained by this invention Natural rubber, a styrene butadiene copolymer, Conjugated diene system elastomers, such as polybutadiene and polyisoprene, A styrene butadiene block copolymer, a styrene isoprene system block copolymer, Styrene system block copolymers, such as those hydrogenation objects, polyethylene, Ethylene propylene rubber, an ethylene propylene diene copolymer, Ethylene system polymers, such as an

ethylene vinyl acetate copolymer and an ethylene ethyl acrylate copolymer. It dissolves with petroleum system hydrocarbon polymers, such as natural polymers, such as polyterpene and a rosin derivative, an aliphatic series system hydrocarbon polymer, and an aromatic series system hydrocarbon polymer, various kinds of waxes, etc.

[0014] In order that the hydrogenation polymer obtained by this invention may show the property excellent in a hue, thermal stability, adhesion, adhesive strength, holding power, etc., it is useful as tackifiers, such as a binder which used various kinds of conjugated diene system elastomers and styrene system block copolymers as the base, and adhesives, and useful as a tackifier especially used for the binder constituent for adhesive tape, such as the Kraft tape, a cellophane tape, and cloth adhesive tape. furthermore, the thing to add in an ethylene vinyl acetate copolymer, these block copolymers, and other thermoplastic polymers, for example, polyethylene, an ethylene ethyl acrylate copolymer, attack tic polypropylene, a polyamide, polyester, etc. -- the object for the object for traffic marking paint, the object for coatings, and printing ink -- it can be used.

[0015]

[Example] An example and the example of reference are given to below, and this invention is explained to it still more concretely. In addition, the section in an example and % are weight criteria as long as there is no notice especially. In addition, the rate of hydrogenation was 60MHz NMR, softening temperature is JIS K-2531 (ring and ball method), and the Gardner hue was measured according to ASTM D-154-58. Moreover, each physical properties of the example of reference were measured by the following approaches.

Tuck :P STC#6 The shot which has a 7/16 inch diameter for a with a rolling ball tuck method include angle of 21.5 degrees ramp top is rolled, and the distance which stops on the tape sample stretched on the horizontal plate is found. However, the inlet length could be 15cm when measuring in a 10-degree C environment.

Adhesive strength: Cut a JIS Z1522 180" friction test adhesive tape sample to 10cm width, paste SUS304 plate, and exfoliate in tensile strength 300 mm/min.

[0016] Holding power: A JIS Z1524 adhesive-tape sample is cut to 15mm width, paste SUS304 plate by regularity area 15mmx25mm, hang 1.2kg of loads at the end of a tape, and find time amount until an adhesion side separates and falls. However, environmental temperature was performed at 40 degrees C.

After carrying out the nitrogen purge of the inside of the dicyclopentadiene 70 weight section, the 5-methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en (compound A) 30 weight section and xylene 200 weight section preparation, and a system to the autoclave equipped with example 1 agitator, it reacted by temperature 260 \*\* for 6 hours. Reaction mixture was applied to vacuum distillation, low boiling materials, such as an unreacted raw material, a low-grade polymer, and a xylene, were distilled off, and the polymer-like matter with transparent light yellow was obtained. This polymer-like matter 50 section was dissolved in the tetrahydrofuran, it taught again the autoclave, the 5% palladium / carbon catalyst 2 section was added, and the hydrogenation reaction was performed at the hydrogen pressure of 50kg/cm<sup>2</sup>, and the temperature of 200 degrees C for 6 hours. After purging the hydrogen after a reaction, the reaction solution was taken out, the reaction solution after filtration was covered over distillation for the catalyst, the solvent was distilled off, and the hydrogenation polymer was obtained. The rate of hydrogenation was about 100%. The softening temperature of this polymer and a hue were measured. The measurement result was shown in Table 1.

[0017] example 2 raw material -- the mixture 50 weight section [ of 6.5.1.13, 6.02, 7.09, 13] pen TADEKA -4, 11-diene, and 7.4.0.02, 7.13 and PENTA cyclo [6.110, 13] pen TADEKA -4, and 11-diene ], and PENTA cyclo [5-methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en 50 weight section The used outside experimented like the example 1. The evaluation result of the obtained hydrogenation polymer was shown in Table 1.

example 3 raw material -- the dicyclopentadiene 60 weight section, the 5-methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en 30 weight section, and 8-methyl-8-methoxycarbonyl tetracyclo [-- 4. -- except for having used 4.0.12, 5.17, and the 10]-3-dodecen (compound B) 10 weight section -- example 1 It experimented similarly and the hydrogenation polymer was

obtained. The evaluation result of the obtained hydrogenation polymer was shown in Table 1. the autoclave equipped with example 4 agitator -- 8-methyl-8-methoxycarbonyl tetracyclo [--- 4. --- after teaching 4.0.12, 5.17, and the 10]-3-dodecen 100 weight section and the xylene 200 weight section and carrying out the nitrogen purge of the inside of a system, it reacted at the temperature of 260 degrees C for 6 hours. Reaction mixture was applied to vacuum distillation, low-boiling point objects, such as an unreacted raw material low-grade polymer and a xylene, were distilled, and the polymer-like matter with transparent light yellow was obtained. This polymer-like matter was hydrogenated like the example 1, and the hydrogenation polymer was obtained. The rate of hydrogenation was about 100%. The evaluation result of the obtained polymer was shown in Table -1.

[0018] The dicyclopentadiene 1000 weight section and the methyl-methacrylate 500 weight section were taught to the autoclave equipped with example 5 agitator, and it reacted to it at 180 degrees C for 5 hours. after distilling off a small amount of cyclopentadiene generated by decomposition of an unreacted methyl methacrylate, a dicyclopentadiene, and a dicyclopentadiene by distilling reaction mixture, and the generated tricyclo pentadiene -- further -- a degree of vacuum -- raising -- 8-methyl-8-methoxycarbonyl tetracyclo [--- 4. --- 4.0.12, 5.17, and 10]-3-dodecen were distilled off. The still residue of distillation the analyzed result -- little 8-methyl-8-methoxycarbonyl tetracyclo [--- 4. --- 4.0.12, 5.17, and 10] -- except for -3-dodecen -- 40 % of the weight of tetramers of a cyclopentadiene, and 11-methyl-11-methoxycarbonyl hexa cyclo [--- 6. --- 6.1. --- 13, 6.110, 13.02, 7.09, and 42 % of the weight (compound C) of 14]-4-heptadecene -- and the matter of a high-boiling point existed further. This still residue 60 weight section and dicyclopentadiene were taught to the autoclave, and 40 weight sections and the xylene 200 weight section were reacted at 280 degrees C in them for 4 hours. The low boiling material was distilled off by the distillation after a reaction, and the quality of polymer choice goods was obtained. After dissolving the obtained polymer-like matter 50 weight section in the tetrahydrofuran 450 weight section, it taught again the autoclave, 10 weight sections were added for the stabilization nickel catalyst (product made from the JGC chemistry), and the hydrogenation reaction was performed at 230 degrees C at 100kg/cm2 for 4 hours. The catalyst and the solvent were removed from the hydrogenation reaction solution, and 145 degrees C of softening temperatures and the polymer of Gardner hue <1 were obtained. In addition, the rate of hydrogenation was about 100%.

[0019] The binder constituent which flows uniformly and smoothly was obtained by putting into the lab PURASUTO mill which heated the hydrogenation polymer 120 weight section obtained in the example of reference 1 example 1, the styrene isoprene system block polymer (JSR SIS5000) 100 weight section, the process oil (Idemitsu Kosan Guyana process oil NM-280) 20 weight section, and the antioxidant (3 Japanese Federation of Chemical Industry Workers' Unions Sun Serra - PZ) 4 weight section at 140 degrees C, and mixing for 30 minutes. This binder constituent was taken out in the state of melting, it coated so that it might become the thickness of 35micro on polyester film by the applicator, and the adhesive tape sample was created. each physical-properties constituent measurement of the tuck (10 degrees C) about this adhesive tape sample, adhesion (23 degrees C), and holding power (40 degrees C) -- it carried out. The result was shown in Table 2.

The same experiment as the example 1 of reference was conducted except having changed the example 2 of reference - 5 hydrogenation polymer into the petroleum polymer obtained in the examples 2-5, respectively. The result was shown in Table 2.

The same experiment as the example 1 of reference was conducted using the petroleum polymer obtained in the example 1 of a comparison in the example 1 of comparison reference - 2 hydrogenation polymer, and the commercial petroleum polymer ( S KORETTSU 1310 made from Exxon chemistry). It is a table about a result. It was shown in 2.

[0020] The hydrogenation polymer 100 weight section, the ethylene vinyl acetate copolymer (2% of vinyl acetate) which were obtained in the example of reference 5 example 1 Melt index 150g / 10 minutes, the Mitsui poly chemical \*\* The 100 weight sections, Carry out the seal of other aluminum plates for 2 seconds in piles to the bottom of a condition with a pressure [ of 1.0kg/cm2 ], and a temperature of 150 degrees C after making the compound which consists of



the paraffin wax 40 section, carrying out melting mixing of this at 180 degrees C and applying the mixture to the aluminum plate of 0.1mm thickness at 0.2mm \*\*. It welded and the laminating sample was made. This sample was cut in the shape of [ of 25mm width ] a strip of paper, the test piece was pulled the rate for 200mm/as a test piece in each temperature of 10 degrees C, 20 degrees C, 30 degrees C, and 40 degrees C, and the adhesive strength by the exfoliating method was measured 90 degrees. The adhesive strength in each temperature was 500, 700, and 1450 or 1300g/inch, respectively.

[Effect of the Invention] The hydrogenation polymer which demonstrates the effectiveness which was excellent as a tackifier of various adhesives or a binder by the manufacture approach of this invention can be obtained. That is, the hydrogenation polymer by this invention shows the property excellent in thermal stability, tackiness, adhesive strength, holding power, etc., and the thermal resistance which was excellent in especially the hot melt constituent has it.

[Table 1]

	実施例 1	実施例 2	実施例 3	実施例 4	比較例 1
C P D系化合物 (種類、仕込量)	DCP 70	TCP 50	DCP 60	-	DCP 100
一般式(1)化合物* (種類、仕込量)	化合物 A 30	化合物 A 50	化合物 A 30 化合物 B 10	化合物 B 100	-
軟化点 (℃)	115	135	125	140	160
分子量	1300	1460	1400	1300	2400
分子量>5000の含有率(%)	0.4	0.2	0.4	0.2	4.8
ガードナー色相	<1	<1	<1	<1	<1

\*:化合物 A : 5-メチル-5-メトキシカルボニルピシクロ [2, 2, 1] ヘプト-2-エン  
化合物 B : 8-メチル-8-メトキシカルボニルピシクロ [4, 4, 0, 1<sup>2,10</sup>, 1<sup>7,10</sup>] -3-ノドアセン

[Table 2]

第2表

	参考例1	参考例2	参考例3	参考例4	参考例5	比較参考例1	比較参考例2
粘着剤樹脂	実施例1の樹脂	実施例2の樹脂	実施例3の樹脂	実施例4の樹脂	実施例5の樹脂	比較例1の樹脂	市販樹脂(○1)
高温保持力	148	180	160	210	230	160	180
低温タック	2	2	2	4	6	>25	>25
接着力	1080	1100	1060	1100	1200	800	1100

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TECHNICAL FIELD

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[Industrial Application] This invention relates to the manufacture approach of a new hydrogenation polymer.

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PRIOR ART

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[Description of the Prior Art] Generally hydrogenation polymers, such as a hydride of the poly cyclopentadiene, are used as a tackifier for hot melt mold adhesives which makes a base polymer the polymer which hydrogenated the diene section of a styrene butadiene block copolymer, a styrene isoprene block copolymer, and these polymers.

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EFFECT OF THE INVENTION

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[Effect of the Invention] The hydrogenation polymer which demonstrates the effectiveness which was excellent as a tackifier of various adhesives or a binder by the manufacture approach of this invention can be obtained. That is, the hydrogenation polymer by this invention shows the property excellent in thermal stability, tackiness, adhesive strength, holding power, etc., and the thermal resistance which was excellent in especially the hot melt constituent has it.

[Table 1]

第1表

[Table 2]

	実施例 1	実施例 2	実施例 3	実施例 4	比較例 1
C/P/D系化合物 (種類、仕込量)	DCP 70	TCP 50	DCP 60	-	DCP 100
一粒子(1)化合物* (種類、仕込量)	化合物A 30	化合物A 50	化合物A 30 化合物B 10	化合物B 100	-
軟化点 (°C)	115	135	125	140	160
分子量	1300	1460	1400	1300	2400
分子量>5000の含有率(%)	0.4	0.2	0.4	0.2	4.8
ガードナー色相	<1	<1	<1	<1	<1

\*：化合物A：5-メチル-5-メトキシカルボニルビシクロ[2.2.1]ヘプト-2-エン

化合物B：8-メチル-8-メトキシカルボニルチトラシクロ[4.4.0.1<sup>2,5</sup>.1<sup>7,10</sup>]-3-ノドアセン

第2表

	参考例1	参考例2	参考例3	参考例4	参考例5	比較参考例1	比較参考例2
粘着剤樹脂	実施例1の樹脂	実施例2の樹脂	実施例3の樹脂	実施例4の樹脂	実施例5の樹脂	比較例1の樹脂	市販樹脂(※1)
高温保持力	148	180	160	210	230	160	180
低温タック	2	2	2	4	6	>25	>25
接着力	1080	1100	1060	1100	1200	800	1100

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TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention] However, compared with organic solvent mold adhesive tape, the adhesive strength and the low-temperature tuck at the time of an elevated temperature were insufficient for the hot melt mold binder which made the conventional hydrogenation polymer the tackifier, and development of the tackifier from which the hot melt mold binder of high performance is obtained more was desired.

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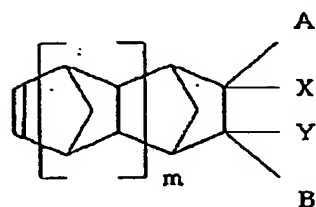
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## MEANS

[Means for Solving the Problem] The purpose of this invention is to offer the manufacturing method of the hydrogenation polymer excellent in the hue and the adhesion property. That is, this invention carries out the polymerization of the 10 - 100 % of the weight of the norbornene system compounds shown by 0 - 90 % of the weight of cyclopentadiene system monomers, and the general formula (I), and offers the manufacture approach of the hydrogenation polymer characterized by subsequently hydrogenating the obtained polymer under existence of a hydrogenation catalyst.

[0005]

[Formula 2]



[-- for the inside A and B of a formula, it is a hydrogen atom or the hydrocarbon group of carbon numbers 1-10, and X and Y are a hydrogen atom or the organic radical of monovalence, at least one of X and the Y shows the radical which has polarities other than a hydrogen atom and a hydrocarbon group, and m is 0-2.] Hereafter, the component of this invention is explained in full detail. As a cyclopentadiene system monomer used in this invention, lower order Diels-Alder adducts, such as \*\*\*\*\* of those \*\*\*\*\*, \*\*\*\*\*, \*\*\*\*\*, and a cyclopentadiene (alkylation), and dienes, such as an isoprene and piperylene, etc. can be mentioned to the dimer of an others, a cyclopentadiene, or its alkylation object, a trimer, a tetramer, and a pan.

[ cyclopentadiene / like a cyclopentadiene, a methyl cyclopentadiene, an ethyl cyclopentadiene, and a dimethyl cyclopentadiene / low-grade alkylation ]

[0006] Moreover, a part of cyclopentadiene system monomer can also be replaced with the ester of alpha, such as dienes, such as styrene system hydrocarbon compounds, such as an indene, a methyl indene, cumarone, etc. and styrene, alpha-methyl styrene, a butadiene, and an isoprene, a methyl acrylate, and a methyl methacrylate, and beta unsaturated carboxylic acid etc. As a norbornene system compound shown by the general formula (I) which can be used in this invention A 5-methoxycarbonyl bicyclo [2.2.1] hept-2-en, A 5-butoxycarbonyl bicyclo [2.2.1] hept-2-en, A 5-cyclohexyloxy carbonyl bicyclo [2.2.1] hept-2-en, A 5-(4'-t-butyl cyclohexyloxy) carbonyl bicyclo [2.2.1] hept-2-en, 5 - Methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en, 5 - Methyl-5-(1-methyl propoxy) carbonyl bicyclo [2.2.1] hept-2-en, A 5-methyl-5-phenoxy carbonyl bicyclo [2.2.1] hept-2-en, 8-methoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 4.4.0.12, 5.17, 8-(1-methyl propoxy) carbonyl tetracyclo [10]-3-dodecen, 8-phenoxy carbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3 - Dodecen, 8-methyl-8-methoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-ethoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-isopropoxycarbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-cyclohexyloxy carbonyl tetracyclo [4. 4.0.12, 5.17, 10]-3-dodecen, 8-methyl-8-(4'-t-butyl

cyclohexyloxy) carbonyl tetra... [--- 4. --- 4.0.12, 5.17, 10]-3-dodece... and 8-cyano tetracyclo [--- 4. --- 4.0.12, 5.17, 10]-3-dodecen, etc. can be mentioned.

[0007] The hydrogenation polymers obtained as a radical which has a polarity in the norbornene system compound expressed with the general formula (I) used in this invention are an elevated temperature, high adhesive strength, and the point of having low hygroscopicity, the radical expressed with formula-(CH<sub>2</sub>)<sub>n</sub>COOR<sub>1</sub> is desirable, and this radical is desirable because the hygroscopicity of the hydrogenation polymer with which it is acquired that it is one per norbornene compound 1 molecule expressed with (a general formula I) becomes low. Moreover, n among the radicals expressed with formula-(CH<sub>2</sub>)<sub>n</sub>COOR<sub>1</sub> That it is desirable since the thermal stability of the hydrogenation polymer with which the thing which has a small value is obtained becomes high, and it is n=0 usually The hydrogenation polymer obtained when compounding a monomer is desirable at the point that a good property is acquired. the hygroscopicity of a hydrogenation polymer becomes small so that a carbon number increases, although R<sub>1</sub> is the hydrocarbon group of carbon numbers 1-20, but since softening temperature and thermal stability fall, the point of balance with the softening temperature of a hydrogenation polymer and the hygroscopicity which are acquired to R<sub>1</sub> is the chain-like hydrocarbon group of carbon numbers 1-4, or the cyclic-hydrocarbon radical of carbon numbers 5-7 -- it is desirable.

[0008] Furthermore, it is desirable in order that that by which the hydrocarbon group of carbon numbers 1-10, especially the methyl group are permuted by the carbon atom which the radical expressed with formula-(CH<sub>2</sub>)<sub>n</sub>COOR<sub>1</sub> combined may reduce hygroscopicity, without reducing the softening temperature of the hydrogenation polymer obtained. The ratio of the cyclopentadiene system monomer in monomer mixture is 0 - 60% of the weight of the range more preferably zero to 80% of the weight zero to 90% of the weight. If this ratio exceeds 90 % of the weight, when it uses as a tackifier of a hot melt constituent, the hydride of the polymer obtained will not be able to give sufficient cold resistance, and will be inferior also in the adhesive property at the time of an elevated temperature. The ratio of the norbornene system compound shown by the general formula in monomer mixture (I) is 10 - 100 % of the weight, and is 40 - 100 % of the weight more preferably 20 to 100% of the weight. If this ratio is less than 10 % of the weight, when that hydride is used for a hot melt constituent, it becomes impossible to demonstrate that unique adhesiveness, and is not desirable.

[0009] In this invention, the polymerization with the norbornene system compound shown by the cyclopentadiene system monomer and the general formula (I) can be performed under existence of a solvent or un-existing by various kinds of approaches, such as cationic polymerization, anionic polymerization, ion coordination polymerization, a radical polymerization, and non-catalyst thermal polymerization. In a cationic polymerization catalyst, for example, a cationic polymerization nature catalyst No. [ 10 ] given [ 16th-volume ] in the petrochemical meeting magazine 865-867th page (1973), Specifically AlCl<sub>3</sub>, AlBr<sub>3</sub>, BF<sub>3</sub>, SnCl<sub>4</sub>, SbCl<sub>5</sub>, FeCl<sub>3</sub>, AlRCl<sub>2</sub> (R: alkyl group), There is AlEt<sub>3</sub>-H<sub>2</sub>O, CCl<sub>3</sub>COOH, and H<sub>2</sub>SO<sub>4</sub> etc., and in these, the yield of a polymer is high, a hue is good, and Lewis acid, especially AlCl<sub>3</sub>, AlBr<sub>3</sub>, etc. are excellent in the point that the high thing of softening temperature is obtained. Moreover, complexes, such as these Lewis acid and alcohol, ester, the ether, and alkyl halide, can also be used suitably.

[0010] For an ion coordination polymerization catalyst, on a Ziegler type catalyst No. [ 8 ] given [ for example, / 16th-volume ] in the petrochemical meeting magazine 699-704th page (1973), and a concrete target The halide of Ti, V, Cr, and Zr, beta-diketone salt, an alcoholate, etc. AlR<sub>3</sub>, AlR<sub>n</sub>X<sub>3-n</sub>, LiR, a MgRX[R:alkyl group, X: There is combination with a halogen atom, n:1 or 2], LiH, etc., and in these, since reactivity is high, the halide of Ti or V and its combination of alkylaluminum are desirable. According to radical polymerization catalyst No. [ 10 ] given [ for example, / 16th-volume ] in the petrochemical meeting magazine 867-871st page (1973), and the polymerization temperature to which specifically has redox system catalysts, such as azo compounds, such as peroxides, such as benzoyl peroxide and tertiary butyl high dollar peroxide, and azo-isobutyro-dinitrile, the hydrogen-peroxide-1st iron compound, and potassium persulfate, and these radical polymerization catalyst was set, it is suitably chosen as radical polymerization catalyst from these.

[0011] Although it can apply if a lot of [ the so-called metathesis ring opening polymerization ]

molecular weight modifiers (for example, 1-hexane etc.) are not added in the polymerization in this invention, and there is nothing that there is nothing, since it is generally easy to give the amount polymer of macromolecules, it is not desirable. In this invention, especially the thing done for thermal polymerization at 200-300 degrees C is desirable under un-existing [ of a radical initiator ] among these polymerizations. In this case, 230-280 degrees C of polymerization temperature can be preferably acquired by carrying out thermal polymerization at 240-270 degrees C more preferably. In this case, polymerization time amount is 0.5 - 20 hours. The molecular weight of the polymer obtained is polystyrene conversion, 300-3000 are desirable, it is desirable that the thing of 5000 or more molecular weight does not contain 5% of the weight or more especially, and the thing of 5000 or more molecular weight is 2 or less % of the weight more preferably. In this invention, the hydrogenation reaction of this polymer is performed by the usual approach. What is used for the hydrogenation reaction of the usual olefin nature compound can be used for the catalyst used at this hydrogenation reaction.

[0012] For example, as a heterogeneous catalyst, the solid-state catalyst which made support, such as carbon, a silica, an alumina, and a titania, support catalyst matter, such as palladium, platinum, a ruthenium, a rhodium, and nickel, is mentioned. The catalyst which supported especially palladium given in JP,3-174406,A to silica magnesia support is activity and a life. It is desirable from points, such as cost, and when a heterogeneous catalyst is used, it can react to a suspension floor and the solid-state floor. Moreover, the catalyst which combined organic lithiums, such as organic aluminum, such as organic solvent soluble nickel [ , such as naphthenic-acid nickel, titanocene dichloride, and cobalt acetylacetonate, ], cobalt, titanium, vanadium compound and triethylaluminum, triisobutylaluminum, and diethyl aluminum mono-chloride, or butyl lithium, as a homogeneous catalyst can be used, and noble-metals complex compound catalysts, such as a chloro tris (triphenyl phosphine) rhodium, can also be used. a hydrogenation reaction -- ordinary pressure - 300 atmospheric pressure -- desirable -- the bottom of the hydrogen gas ambient atmosphere of three to 150 atmospheric pressure, and temperature -- 0-degreeC-300-degreeC -- it can carry out by 20-degreeC-250-degreeC preferably.

[0013] According to this invention, the hydrogenation polymer which has the outstanding hue, i.e., an one or less Gardner color standards [ which were specified to JIS K-5400 ] hue, and has the softening temperature specified to JIS K-2531 [ 60-200-degree C ] is obtained. The hydrogenation polymer obtained by this invention is meltable to aliphatic hydrocarbon, such as a pentane, a hexane, benzene, toluene, a xylene, chloroform, and a carbon tetrachloride, aromatic hydrocarbon, or halogenated hydrocarbon, and is a polymer-like polymer in which low melt viscosity is shown. The hydrogenation polymer obtained by this invention Natural rubber, a styrene butadiene copolymer, Conjugated diene system elastomers, such as polybutadiene and polyisoprene, A styrene butadiene block copolymer, a styrene isoprene system block copolymer, Styrene system block copolymers, such as those hydrogenation objects, polyethylene, Ethylene propylene rubber, an ethylene propylene diene copolymer, Ethylene system polymers, such as an ethylene vinyl acetate copolymer and an ethylene ethyl acrylate copolymer, It dissolves with petroleum system hydrocarbon polymers, such as natural polymers, such as polyterpene and a rosin derivative, an aliphatic series system hydrocarbon polymer, and an aromatic series system hydrocarbon polymer, various kinds of waxes, etc.

[0014] In order that the hydrogenation polymer obtained by this invention may show the property excellent in a hue, thermal stability, adhesion, adhesive strength, holding power, etc., it is useful as tackifiers, such as a binder which used various kinds of conjugated diene system elastomers and styrene system block copolymers as the base, and adhesives, and useful as a tackifier especially used for the binder constituent for adhesive tape, such as the Kraft tape, a cellophane tape, and cloth adhesive tape. furthermore, the thing to add in an ethylene vinyl acetate copolymer, these block copolymers, and other thermoplastic polymers, for example, polyethylene, an ethylene ethyl acrylate copolymer, attack tic polypropylene, a polyamide, polyester, etc. -- the object for the object for traffic marking paint, the object for coatings, and printing ink -- it can be used .

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## EXAMPLE

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[Example] An example and the example of reference are given to below, and this invention is explained to it still more concretely. In addition, the section in an example and % are weight criteria as long as there is no notice especially. In addition, the rate of hydrogenation was 60MHz NMR, softening temperature is JIS K-2531 (ring and ball method), and the Gardner hue was measured according to ASTM D-154-58. Moreover, each physical properties of the example of reference were measured by the following approaches.

Tuck :P STC#6 The shot which has a 7/16 inch diameter for a with a rolling ball tuck method include angle of 21.5 degrees ramp top is rolled, and the distance which stops on the tape sample stretched on the horizontal plate is found. However, the inlet length could be 15cm when measuring in a 10-degree C environment.

Adhesive strength: Cut a JIS Z1522 180" friction test adhesive tape sample to 10cm width, paste SUS304 plate, and exfoliate in tensile strength 300 mm/min.

[0016] Holding power: A JIS Z1524 adhesive-tape sample is cut to 15mm width, paste SUS304 plate by regularity area 15mmx25mm, hang 1.2kg of loads at the end of a tape, and find time amount until an adhesion side separates and falls. However, environmental temperature was performed at 40 degrees C.

After carrying out the nitrogen purge of the inside of the dicyclopentadiene 70 weight section, the 5-methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en (compound A) 30 weight section and xylene 200 weight section preparation, and a system to the autoclave equipped with example 1 agitator, it reacted by temperature 260 \*\* for 6 hours. Reaction mixture was applied to vacuum distillation, low boiling materials, such as an unreacted raw material, a low-grade polymer, and a xylene, were distilled off, and the polymer-like matter with transparent light yellow was obtained. This polymer-like matter 50 section was dissolved in the tetrahydrofuran, it taught again the autoclave, the 5% palladium / carbon catalyst 2 section was added, and the hydrogenation reaction was performed at the hydrogen pressure of 50kg/cm<sup>2</sup>, and the temperature of 200 degrees C for 6 hours. After purging the hydrogen after a reaction, the reaction solution was taken out, the reaction solution after filtration was covered over distillation for the catalyst, the solvent was distilled off, and the hydrogenation polymer was obtained. The rate of hydrogenation was about 100%. The softening temperature of this polymer and a hue were measured. The measurement result was shown in Table 1.

[0017] example 2 raw material -- the mixture 50 weight section [ of 6.5.1.13, 6.02, 7.09, 13] pen TADEKA -4, 11-diene, and 7.4.0.02, 7.13 and PENTA cyclo [6.110, 13] pen TADEKA -4, and 11-diene ], and PENTA cyclo [5-methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en 50 weight section The used outside experimented like the example 1. The evaluation result of the obtained hydrogenation polymer was shown in Table 1.

example 3 raw material -- the dicyclopentadiene 60 weight section, the 5-methyl-5-methoxycarbonyl bicyclo [2.2.1] hept-2-en 30 weight section, and 8-methyl-8-methoxycarbonyl tetracyclo [-- 4. -- except for having used 4.0.12, 5.17, and the 10]-3-dodecen (compound B) 10 weight section -- example 1 It experimented similarly and the hydrogenation polymer was obtained. The evaluation result of the obtained hydrogenation polymer was shown in Table 1. the autoclave equipped with example 4 agitator -- 8-methyl-8-methoxycarbonyl tetracyclo [--

4. -- after teaching 4.0.12, 5.17, and the 10]-3-dodecen 100 weight section and the xylene 200 weight section and carrying out the nitrogen purge of the inside of a system, it reacted at the temperature of 260 degrees C for 6 hours. Reaction mixture was applied to vacuum distillation, low-boiling point objects, such as an unreacted raw material low-grade polymer and a xylene, were distilled, and the polymer-like matter with transparent light yellow was obtained. This polymer-like matter was hydrogenated like the example 1, and the hydrogenation polymer was obtained. The rate of hydrogenation was about 100%. The evaluation result of the obtained polymer was shown in Table -1.

[0018] The dicyclopentadiene 1000 weight section and the methyl-methacrylate 500 weight section were taught to the autoclave equipped with example 5 agitator, and it reacted to it at 180 degrees C for 5 hours. after distilling off a small amount of cyclopentadiene generated by decomposition of an unreacted methyl methacrylate, a dicyclopentadiene, and a dicyclopentadiene by distilling reaction mixture, and the generated tricyclo pentadiene -- further -- a degree of vacuum -- raising -- 8-methyl-8-methoxycarbonyl tetracyclo [-- 4. -- 4.0.12, 5.17, and 10]-3-dodecen were distilled off. The still residue of distillation the analyzed result -- little 8-methyl-8-methoxycarbonyl tetracyclo [-- 4. -- 4.0.12, 5.17, and 10] -- except for -3-dodecen -- 40 % of the weight of tetramers of a cyclopentadiene, and 11-methyl-11-methoxycarbonyl hexa cyclo [-- 6. -- 6.1. -- 13, 6.110, 13.02, 7.09, and 42 % of the weight (compound C) of 14]-4-heptadecene -- and the matter of a high-boiling point existed further. This still residue 60 weight section and dicyclopentadiene were taught to the autoclave, and 40 weight sections and the xylene 200 weight section were reacted at 280 degrees C in them for 4 hours. The low boiling material was distilled off by the distillation after a reaction, and the quality of polymer choice goods was obtained. After dissolving the obtained polymer-like matter 50 weight section in the tetrahydrofuran 450 weight section, it taught again the autoclave, 10 weight sections were added for the stabilization nickel catalyst (product made from the JGC chemistry), and the hydrogenation reaction was performed at 230 degrees C at 100kg/cm2 for 4 hours. The catalyst and the solvent were removed from the hydrogenation reaction solution, and 145 degrees C of softening temperatures and the polymer of Gardner hue <1 were obtained. In addition, the rate of hydrogenation was about 100%.

[0019] The binder constituent which flows uniformly and smoothly was obtained by putting into the lab PURASUTO mill which heated the hydrogenation polymer 120 weight section obtained in the example of reference 1 example 1, the styrene isoprene system block polymer (JSR SIS5000) 100 weight section, the process oil (Idemitsu Kosan Guyana process oil NM-280) 20 weight section, and the antioxidant (3 Japanese Federation of Chemical Industry Workers' Unions Sun Serra - PZ) 4 weight section at 140 degrees C, and mixing for 30 minutes. This binder constituent was taken out in the state of melting, it coated so that it might become the thickness of 35micro on polyester film by the applicator, and the adhesive tape sample was created. each physical-properties constituent measurement of the tuck (10 degrees C) about this adhesive tape sample, adhesion (23 degrees C), and holding power (40 degrees C) -- it carried out. The result was shown in Table 2.

The same experiment as the example 1 of reference was conducted except having changed the example 2 of reference - 5 hydrogenation polymer into the petroleum polymer obtained in the examples 2-5, respectively. The result was shown in Table 2.

The same experiment as the example 1 of reference was conducted using the petroleum polymer obtained in the example 1 of a comparison in the example 1 of comparison reference - 2 hydrogenation polymer, and the commercial petroleum polymer ( S KORETTSU 1310 made from Exxon chemistry). It is a table about a result. It was shown in 2.

[0020] The hydrogenation polymer 100 weight section, the ethylene vinyl acetate copolymer (2% of vinyl acetate) which were obtained in the example of reference 5 example 1 Melt index 150g / 10 minutes, the Mitsui poly chemical \*\* The 100 weight sections, Carry out the seal of other aluminum plates for 2 seconds in piles to the bottom of a condition with a pressure [ of 1.0kg/cm2 ], and a temperature of 150 degrees C after making the compound which consists of the paraffin wax 40 section, carrying out melting mixing of this at 180 degrees C and applying the mixture to the aluminum plate of 0.1mm thickness at 0.2mm \*\*. It welded and the laminating

sample was made. This sample was cut in the shape of [ of 25mm width a strip of paper, the test piece was pulled the rate for 200mm/as a test piece in each temperature of 10 degrees C, 20 degrees C, 30 degrees C, and 40 degrees C, and the adhesive strength by the exfoliating method was measured 90 degrees. The adhesive strength in each temperature was 500, 700, and 1450 or 1300g/inch, respectively.

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